

Jun 2000

Naval Health Research Center Detachment (Toxicology)

**ESTIMATION OF ALUMINUM CONTRIBUTIONS  
OF U.S. NAVY FLIGHT TRAINING OPERATIONS  
IN THE CHESAPEAKE BAY**

CODY L. WILSON  
ANIS MILADI  
ROBERT L. CARPENTER  
WILLIAM K. ALEXANDER  
KENNETH R. STILL

REPORT NO.

TOXDET-00-04

Approved for public release; distribution is unlimited

**NAVAL HEALTH RESEARCH CENTER DETACHMENT (TOXICOLOGY)  
2612 FIFTH STREET, BUILDING 433, AREA B  
WRIGHT-PATTERSON AIR FORCE BASE, OHIO 45433-7903**



QUALITY IMPROVED 4  
20010216 006



# **Estimation of Aluminum Contributions of U.S. Navy Flight Training Operations in the Chesapeake Bay**

**Cody L. Wilson, Ph.D.  
LT, MSC, USNR  
NHRC/TD**

**Anis Miladi, B.S.  
ENS, MC, USNR-R  
NHRC/TD**

**Robert L. Carpenter, Ph.D.  
Senior Scientist  
NHRC/TD**

**William K. Alexander, M.S.  
CDR, MSC, USN  
NAMRU-2**

**Kenneth R. Still, Ph.D.  
CAPT, MSC, USN  
Officer-in-Charge  
NHRC/TD**

Naval Health Research Center Detachment (Toxicology)  
2612 Fifth Street, Bldg 433, Area B  
Wright-Patterson Air Force Base, Ohio 45433-7903

Report No. TOXDET-00-04 was supported by the Naval Health Research Center, Department of the Navy, under Work Unit No. 63706N-M00095.004.1822. The views expressed in this article are those of the authors and do not reflect the official policy or position of the Department of Defense, nor the U.S. Government. Approved for public release, distribution unlimited.

## **PREFACE**

This document reports the results from an investigation of the impact of aluminized glass chaff countermeasures on environmental aluminum levels in the Chesapeake Bay. This study was conducted by the Naval Health Research Detachment (Toxicology) in response to concerns expressed over the potential environmental hazards that might be associated with the release of aluminized glass chaff fibers during training exercises by Naval aviators. This work was sponsored by the Naval Air Warfare Center/Aircraft Division under Work Unit # 63706N-M00095.004.1822 and was performed under the direction of CAPT Kenneth R. Still, MSC, USN, Officer-in-Charge NHRC/TD.

The opinions contained herein are those of the authors and are not to be construed as official or reflecting the view of the Department of the Navy or the Naval Services at large.

## TABLE OF CONTENTS

SECTION	PAGE
Executive Summary .....	v
Abstract .....	vi
List of Abbreviations .....	vii
List of Tables .....	viii
List of Figures .....	ix
Introduction.....	1
Experimental.....	3
Results.....	5
Conclusions.....	11
References.....	13
Table 1 .....	16
Figures 1 - 4 .....	17
Table 2 .....	21

## **EXECUTIVE SUMMARY**

### **PROBLEM**

Chaff used to provide protection against radar based attack on aircraft and other military vehicles is composed of aluminum coated glass fibers. Concern has been expressed as to the environmental hazard and potential for human health risk associated with routine release of this material during training exercises.

### **OBJECTIVE**

The objective of this study was to evaluate the impact of U.S. Navy flight training operations on aluminum content in the Chesapeake Beach region of the Chesapeake Bay, an area over which chaff countermeasure flight training operations have been conducted for nearly a quarter century.

### **RESULTS**

Exchangeable and monomeric aluminum content in sediment from the flight path within the Bay is not significantly different from nearby background levels within the Bay. Background residential exchangeable aluminum levels were not significantly different from soil samples obtained from a residential area adjacent to the NRL-CBD complex at Chesapeake Beach.

### **CONCLUSION**

Deployment of aluminized glass chaff countermeasures over the NRL-CBD Chesapeake Bay area appears to have resulted in a minimal, but not statistically significant, increase in bay sediment exchangeable and monomeric aluminum. Residential soil EA levels are several orders of magnitude lower than the current EPA Region III limit for total aluminum in residential soil. Within the context of aluminum ingestion by humans, in comparison with total aluminum content of several commonly consumed foods, exchangeable and total monomeric aluminum fall below or within the range of these values. Solution ions and other factors shown to be important in Al solubility and subsequent bioavailability are either below median levels as reported in other studies or do not appear to significantly alter Al solubility. These data suggest that current and estimated use of aluminized chaff by American forces worldwide will not raise total aluminum levels to EPA Screening Levels.

## **ABSTRACT**

The U. S. Navy currently uses aluminized glass chaff as a passive countermeasure for radar-guided threats to aircraft and surface ships. Chaff countermeasures are composed primarily of aluminized glass strands with lengths of up to 2 inches and a diameter of approximately 1 mil. Until recently, research and development efforts and production lot testing of chaff countermeasures have been conducted primarily at the Naval Research Laboratory—Chesapeake Bay Detachment (NRL-CBD) near Chesapeake Beach, MD. Chaff has been deployed in training and production lot testing in this region of the Bay for nearly 25 years, suggesting a potentially significant contribution to soil and sediment aluminum levels. This study examined the exchangeable and monomeric aluminum content of bay sediment and soil from a nearby residential area to address concerns over the environmental and potential human health impact of deployed chaff countermeasures. The results suggest that sediment monomeric and exchangeable aluminum levels in the NRL-CBD testing flight path area are not significantly different ( $p < 0.01$ ) from nearby background levels within the Bay. Moreover, background residential EA levels were not significantly different ( $p < 0.01$ ) from soil samples obtained from a residential area adjacent to NRL-CBD.

## **KEY WORDS**

Chaff, exchangeable aluminum, monomeric aluminum, aluminum, sediment, Chesapeake Bay.

## LIST OF ABBREVIATIONS

Note common chemical and measurement abbreviations are not included.

AAA	Anti-aircraft artillery
AD	Alzheimer's Disease
Ali	Inorganic Monomeric Aluminum
Alo	Organic Monomeric Aluminum
Alt	Total Monomeric Aluminum
EA	Exchangeable aluminum
GI	Gastrointestinal
NRL-CBD	Naval Research Laboratory—Chesapeake Bay Detachment
SAM	Surface-to-Air Missile

## LIST OF TABLES

Table 1. Basic constituents of chaff. Derived from Military Specification R-6043b

Table 2. Mean values for monomeric and exchangeable aluminum and pH, % solids, %total organic carbon, and exchangeable aluminum normalized to pH for samples obtained from NRL-CBD.



## **LIST OF FIGURES**

- Figure 1. Sampling locations at NRL-CBD.
- Figure 2. Monomeric aluminum concentration of sediment samples obtained from NRL-CBD.
- Figure 3. Exchangeable aluminum concentration of sediment samples obtained from NRL-CBD.
- Figure 4. Comparison of exchangeable and total monomeric aluminum with total aluminum content of commonly consumed foods.

## INTRODUCTION

Aluminized glass chaff consists of a large number (typically 500,000) of fibers that reflect or mask radar signals and serves as one of a battery of defensive mechanisms employed by military aircraft and ships to avoid adversary defense systems such as surface-to-air missiles (SAMs), anti-aircraft artillery (AAA), and other aircraft. When deployed from aircraft or ships, chaff forms an electromagnetic cloud that temporarily hides the aircraft or ship from radar detection. The use of airborne expendable countermeasures by American military aircraft dates back over fifty years, with the first recorded large-scale use of chaff in combat in 1943 over Bremen, Germany. In the early years, chaff was simply tossed out of airplane windows. Dispersal methods have evolved through spring-loaded or pneumatic machines to the use of pyrotechnic charges, rockets, mortars, airflows, or motors. Today, most aircraft chaff is deployed in small bundles weighing about 170 g and containing several million fibers.

In its original form, chaff consisted of strips of solid aluminum. In the 1980s, this form was replaced by silica glass fibers thinly coated with a metal of high purity (i.e., >99.9%), usually aluminum. Fiber lengths range up to 2 inches and are roughly 1 mil (25  $\mu\text{m}$ ) in diameter. Although primarily used by military aircraft and ships, expendable chaff countermeasures have also found use as obscurants among ground force units. Currently, the services use 70 to 75 locations worldwide as training ranges, with many of the ranges being off-shore. One such training range, the Naval Research Laboratory—Chesapeake Bay Detachment (NRL-CBD) was the site of research and development of Navy radio frequency countermeasures from the mid-1970s to 1995. During this time period, it is estimated that several hundred thousand pounds of aluminized glass chaff were dispersed over this area of the Bay.

Evidence to date suggests that the materials comprising chaff are generally nontoxic except in quantities significantly larger than those any human or animal could be reasonably exposed to from training or combat use. The major components of chaff--silica, aluminum, and stearic acid--are generally found in the environment (Table 1). A member of the common group of silicate minerals, silica (silicon dioxide) is inert in the environment; stearic acid is an 18 carbon saturated fatty acid used as a slip coating to prevent chaff clumping upon deployment and is environmentally degradable. Finally, aluminum is the third most abundant element in and comprises roughly 8% of the earth's crust. Naturally occurring aluminum soil concentrations ranging from 10,000 to 300,000 ppm have been documented by Lindsay and coworkers (1979). This metal has been identified in drinking and surface waters, multiple food products, and pharmaceuticals and humans ingest an estimated 10 mg of this element each day through these sources (USDHHS ATSDR, 1992).

Interestingly, absorption of aluminum by the human gastrointestinal (GI) tract is minimal (<1% of total ingested aluminum (Jouhannau *et al.*, 1997)), with the majority of ingested aluminum accumulating in the intestinal mucosa and subsequently passed in the feces upon mucosal cell turnover (Greger *et al.*, 1983)). Aluminum appears to be poorly absorbed and little evidence of adverse consequences of chronic, low level exposure exists. Despite reported correlations between aluminum deposition in the brain and Alzheimer's disease (AD), the causal link between aluminum exposure and neurodegenerative diseases is tenuous (Crapper-McLachlan *et al.*, 1986; Perl *et al.*, 1982; Shore *et al.*, 1983). Furthermore, the mechanism of aluminum absorption across the GI tract remains elusive. Studies in this laboratory are currently underway to assess the bioavailability of chaff-derived aluminum in the mammalian

gastrointestinal tract. The objective of this investigation was to determine the contribution of chaff countermeasures to environmental levels of bioavailable aluminum.

## **EXPERIMENTAL**

*Environmental Sampling:* Soil and sediment samples were collected from NRL-CBD, near Chesapeake Beach, MD. Sediment samples were obtained using a stainless steel sampling dredge (Lab Safety & Industrial Supplies, Janesville, WI) and stored in No. 23884 Whirl-Pak bags. Access to the bay was obtained in a Boston Whaler watercraft provided by NRL-CBD. Position coordinates (LAT/LON), distance to shore, approximate sampling depth (water column depth for sediment; soil depth for soil), and time of day were recorded for each sample. Distance to shore was measured using a laser rangefinder and LAT/LON were recorded using a Magellan 2000XL global positioning satellite receiver system. Discretionary soil samples were obtained by digging to an eight inch depth using an entrenching tool and samples were taken by precleaned 6 oz. polyethylene scoops and placed into No. 23884 Whirl-Pak bags. Samples were shipped via overnight courier to Columbia Analytical, Inc. (Rochester, NY). Background soil samples were obtained from a vacant lot in a residential area of Prince Frederick, MD. A location in the bay several miles from NRL-CBD provided background sediment samples. The likelihood of deposition of chaff-derived aluminum at this site is remote, and this is discussed further.

*Characterization of Soil and Sediment Samples:* U.S. Environmental Protection Agency (EPA) Water/Wastewater Methods 600 Series were used to determine Total Organic Carbon (TOC; Method # 415), Percent Solids (Method # 160.3), and soil and sediment pH (Method # 150). The

concentration of nine ions ( $\text{Ca}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ , and  $\text{SiO}_2$  (Method # 200.7);  $\text{Cl}^-$  (Method # 325);  $\text{F}^-$  (Method # 340); and  $\text{SO}_4^{2-}$  (Method # 375) was measured in pore waters extracted from sediment samples also using EPA 600 Methods.

*Analysis for Total, Organic, and Inorganic Monomeric Aluminum:* Total monomeric (Alt) and organic monomeric aluminum (Alo) was analyzed by Fitzhugh and colleagues at Syracuse University using a pyrocatechol violet colorimetric technique as described (Driscoll *et al.*, 1984; McAvoy *et al.*, 1992). Briefly, this procedure estimated Alt by formation of an Al-catechol complex at pH 6.2 and colorimetric detection at 590 nm. Alo was determined by passing the sample through an organic exchange column filled with acidic (pH 5) cation exchange resin (Amberlite IR-120). Inorganic monomeric aluminum (Ali) was estimated by subtracting the Alo from the Alt.

*Analysis of Exchangeable Aluminum:* Analysis for exchangeable aluminum (EA) was performed by Columbia Analytical, Inc. according to the method of Cappel and colleagues (Cappel *et al.*, 1987). Briefly, approximately 2.5-g air dried soil or sediment was mixed with 1N KCl, extracted through a 60-cc extraction syringe fitted with a 1-g ball of filter pulp, and the resulting extract was analyzed for EA by inductively-coupled plasma-mass spectrometry (EPA 600 Series Method 200.8).

*Statistical Analysis:* Analyses were carried out in triplicate unless stated otherwise and results are expressed as means  $\pm$ SD. Statistical analysis was performed using Graphed Prism V 3.00.

Students' *t* test was used to compare differences between individual means. The level of significance for all analyses was  $p < 0.01$  unless otherwise indicated.

## RESULTS

Aluminum is the third most abundant element in the earth's crust; however, within the lithosphere, this element is predominately sequestered by aluminosilicate mineral matter and generally unavailable for chemical and biological reactions (Driscoll et al., 1995). Among processes that can contribute to pools of reactively available aluminum are biological assimilation and release (although this is considered a minor pathway), weathering of aluminosilicate minerals, and organic and strong acid dissolution (Driscoll et al., 1995). Hitherto, no requirement for aluminum in plant or animal systems has been identified. Consequently, this element does not generally accumulate in living tissue, but in its bioavailable form Al can be toxic to plants and some animals.

Aluminum concentrations were measured in samples obtained from the NRL-CBD, an area over which deployment of aluminized glass chaff has been tested for nearly 25 years. Figure 1 denotes collection points for 24 sediment samples from a 6.47 km<sup>2</sup> section of the Chesapeake Bay, an area which corresponded to the flight path utilized in chaff deployment exercises.

### Alt, Alo, Ali and EA Concentrations

Sediment samples from the flight path and the background site were centrifuged to extract pore water, from which total aluminum was separated into organic and inorganic monomeric fractions as described in the Experimental section. Alt concentration was determined

for flight path and background bay sediment samples, with no significant difference ( $p < 0.05$ ) between the two groups (Figure 2, Table 2). Concentration of the Ali fraction of flight path sediment was numerically but not statistically different from background. Average Alo concentrations from flight path sediment were not significantly different from bay background (Figure 2, Table 2). Multiple investigators report that organically complexed forms of aluminum in soil solutions and natural waters are much less toxic to plants and aquatic life than  $Al^{3+}$  or its hydrated inorganic monomers even in acidic soils (Driscoll et al., 1980; Thomas et al., 1984; WHO, 1997). Monomeric aluminum values were not derived for soil samples, as sufficient pore water could not be extracted from the samples to perform a reliable analysis.

The sediment solid phase was analyzed for EA, which is the population of aluminum bound to the soil by electrostatic charge, and is generally controlled by the solubility of the non-EA fraction. EA is a good indicator of field acidity and of the concentration of toxic aluminum species present in a medium (Cappo et al., 1987). These results show that EA from flight path bay sediment was not significantly different from background bay sediment (Figure 3, Table 2), and low EA content has been correlated with decreased aluminum toxicity in plants (Hoyt et al., 1975). EA concentrations were also evaluated in soil samples taken from a residential area adjacent to the NRL-CBD complex and a background residential area in Prince Frederick, MD (Table 2). However, EA concentrations were not significantly different between the two groups.

### Effects of Acidity

Speciation, toxicity, and bioavailability of aluminum are closely related to pH and this relationship has been reported by numerous other investigators. In the present study, a relatively strong inverse correlation is observed ( $r = -0.84$ ) between EA concentration and pH, and this

finding is consistent with data from other investigators reporting migration of Al and H ions from colloids to soil solution in low pH environments. When normalized to soil pH, EA in both flight path and background bay sediment was found to be significantly ( $p < 0.05$ ) lower than the background residential site (Table 2). Interestingly, the EPA Region III soil screening level for total aluminum in residential soil is 78,000 mg/kg. The average EA level from the background residential and shore residential soil sites was approximately 1700 and 1300 times lower, respectively, than the limit for total aluminum in residential soil.

#### Potential Complexing Agents

The concentration of several ions shown to be important in aluminum solubility were also measured in sediment pore water extracts. Aluminum forms strong complexes with  $\text{OH}^-$ ,  $\text{F}^-$ , and  $\text{SO}_4^{2-}$  (Roberson et al., 1995). Although circumneutral pH values were observed in both background and flight path sediment, in acidic soils, strong fluoride ( $\text{F}^-$ ) complexes form with  $\text{Al}^{3+}$ , contributing to total soluble aluminum (Lindsey et al., 1995).  $[\text{F}^-]$  was determined to be  $67.19 \pm 19.05 \mu\text{M}$  in pore water extracted from flight path sediment and was not significantly different from the background measurement. Moreover, aluminosilicate complexes have been shown to account for up to 95% of total Al in natural waters (Browne et al., 1992). Silicate concentration averaged  $28.42 \pm 4.13 \text{ mg/L}$  in pore water extracted from flight path sediment and this value did not differ significantly from the background concentration. Median dissolved silicate concentration in surface waters was reported to be  $300 \mu\text{M}$  (Stumm et al., 1970).

$\text{Al}^{3+}$  also forms solution complexes with sulfate which was found to be significantly lower ( $0.8 \pm 0.2$ -fold) in flight path pore water than background (Lindsay et al., 1995). Absolute levels were  $17.34 \pm 0.72$  (background) and  $14.10 \pm 3.48 \text{ mM}$  in pore water extracted from flight



path sediment. Exchangeable and Ali remain relatively low and this finding is consistent with studies demonstrating that Al-SO<sub>4</sub> complexes are significant only at low pH (Stumm et al., 1970).

K<sup>+</sup>, Cl<sup>-</sup>, and Ca<sup>2+</sup> were measured but values for the concentration of these ions in flight path sediment were not significantly different from background. Mn<sup>+</sup> concentration was higher in flight path sediment than background ( $4.25 \pm 3.4$  vs.  $1.26 \pm 0.68$  mg/L). Flight path iron levels were not found to be significantly different from background sediment pore water ( $93.8 \pm 163.9$  vs.  $4.35 \pm 2.32$  mg/L); however, four extremely large values contributed to a relatively high mean concentration in the flight path sediment. It is hypothesized that these iron-rich samples were anthropogenic, as this area is heavily crabbed and fished.

#### Aluminum-related Disease

Initial studies of environmental aluminum exposure as a risk factor for Alzheimer's Disease (AD) began in the 1980s, when reports of the increased level of aluminum in brains of AD patients suggested that this metal might be a factor. Although multiple reports suggest a role for aluminum in the pathogenesis of AD (Crapper-McLachlan et al, 1986; Crapper-McLachlan et al., 1989; Guy et al., 1991; Mesco et al., 1991; Good et al., 1992; Shin et al., 1994), the data are tenuous and the conclusions highly controversial. Of the twenty studies examining the relationship between drinking water aluminum and AD, only four are consistent with a positive relationship between AD and drinking water aluminum; however, these four studies were weakened by lack of adjustment from other confounding or risk factors (Crapper-McLachlan et al., 1986; Martyn et al., 1989; Neri et al., 1991; Flaten et al., 1991). Moreover, recent data

suggests that only 5% of daily aluminum intake is accounted for by drinking water and all of the studies examined drinking water as the sole source of aluminum (WHO, 1997).

#### Chaff Leachate Toxicity

The U.S. Army conducted a chaff fiber fate study by placing up to 1000 mg/L chaff fibers in fresh water of varying hardness and seawater for 21 days. After 21 days in waters of varying hardness or seawater with chaff concentrations ranging up to 1000 mg/L, no toxicity was observed in the 48 h acute toxicity test in Mysid shrimp or Daphnia and the solution was not toxic to Sheepshead minnow in a 96 h study (Haley et al., 1992). Similarly, a U.S. Air Force group reviewed several studies examining the environmental effects of aluminized glass chaff and concluded that aluminum toxicity due to chaff is not a concern in aquatic environments (USAF ACC, 1997).

#### Impact Modeling and Future Use of Chaff

Initiatives between the Department of Defense (DoD) and several Department of Interior (DoI) agencies are aimed at minimizing the impact of chaff on public lands. In agreements with both the Fish and Wildlife Service and Bureau of Land Management, the military services tightly control chaff use over wildlife refuges, Native American reservations, and public lands near military training grounds. A committee comprised of representatives from the Bureau of Land Management and the military services was constituted to evaluate policies on chaff deployment over public lands (GAO, 1998). Moreover, the Navy has limited agreements to restrict chaff use over wildlife refuges and public lands in the interest of protection of sensitive species from potential impacts (GAO, 1998).

In addition to a series of policies governing the use of chaff in training or test and evaluation, the military services have conducted several studies examining the environmental impact of chaff, including one comprehensive evaluation by a Select Panel of academic experts (Hullar et al., 1999). This panel developed several worst case scenarios to calculate a unit area exposure of chaff in two high chaff use regions within the United States (Hullar et al, 1999). These worst case scenarios are based on a number of assumptions, such as no horizontal drift of chaff and that all chaff was released within the military operating area (MOA). Because of its relevancy to the sensitive coastal watershed, the use of chaff at the Patuxent Naval Air Station (NAS), Maryland south of the NRL-CBD complex in the Chesapeake Bay watershed and the current site of chaff production lot testing for the Navy was considered. In this case, the panel reported 683 bundles (136 kg) in 1998 over a MOA of 6216 km<sup>2</sup> resulting in a chaff distribution of 0.02 kg/km<sup>2</sup>. The resultant aluminum concentration is 0.344 mg Al/m<sup>3</sup> soil/y for the top 1" of soil. Using a bulk soil density of 2600 kg/m<sup>3</sup> for clays in calculations, chaff usage at Patuxent NAS would need to continue at the 1998 rate for nearly 600 million years to reach the EPA Region III soil screening level.

In contrast to the previous example where approximate chaff utilization was known, in the present study, potential anthropogenic changes soil and sediment in aluminum concentration were measured in the NRL-CBD region of the Chesapeake Bay. Again, the vast majority of chaff was deployed under highly controlled and calm conditions at low altitude, suggesting the chaff contained in one bundle (190 g; approx. 6 million fibers) might be distributed over the flight path area of 6.47 km<sup>2</sup>. A volume distribution of 0.483 mg Al/m<sup>3</sup> can be calculated for the top 0.0254 m of soil. Back calculation using the Bay Flight Path sediment total monomeric aluminum value

of 1.97 mg/kg (95% CI) (Table 2) results in 2400 to 6400 chaff bundles being released over a 25 year period (again using the range of soil bulk density values of 1000 kg/m<sup>3</sup> to 2600 kg/m<sup>3</sup>). It is clear that chaff deposition in these areas would require time of geologic proportions (nearly 2.6 million years at the 1998 rate of usage) to reach any substantial accumulation (i.e., the EPA Region III soil screening level of 78,000 mg/kg). Even if the majority of chaff was to float on the surface of the water and accumulate on shore, it is evident from soil Al values measured at or near the shoreline that the EPA Region III screening limit would not be exceeded. Although the exact number of chaff bundles released over the 25 year period during which NRL-CBD was active could not be ascertained, the calculated values are lower than estimated use suggesting sediment sampling may not be an accurate means of estimating past chaff usage.

## CONCLUSIONS

Based on this study, deployment of aluminized chaff countermeasures over the NRL-CBD Chesapeake Bay area appears to have resulted in a minimal, but not statistically significant, increase in bay sediment exchangeable and monomeric aluminum. Residential soil EA levels are several orders of magnitude lower than the current EPA Region III limit for total aluminum in residential soil. Within the context of aluminum ingestion by humans, in comparison with total aluminum content of several commonly consumed foods, EA and Alt fall below or within the range of these values (Figure 4). Solution ions and other factors shown to be important in Al solubility and subsequent bioavailability are either below median levels as reported in other studies or do not appear to significantly alter Al solubility. These data suggest that current and

estimated use of aluminized chaff by American forces worldwide will not raise total aluminum levels to EPA Screening Levels.

## REFERENCES

- Adams, F.**, Ed. (1984) Soil Acidity and Liming. Agronomy Series No. 12, American Society of Agronomy, Madison, WI.
- Agency for Toxic Substances and Disease Registry.** (1992) Toxicological Profile for Aluminum. USDHHS ATSDR, TP-91/01.
- Browne, B.A. and Driscoll, C.T.** (1992) Soluble aluminum silicates: stoichiometry, stability, and implications for environmental geochemistry, *Science*, 256, 1667.
- Cappo, K.A., Blume, L.J., Raab, G.A., Bartz, J.K., Engels, J.L.** (1987) Analytical Methods Manual for the Direct/Delayed Response Project Soil Survey. Environmental Monitoring Systems Laboratory, Office of Research and Development, USEPA 600/8-87/020.
- Crapper-McLachlan, D.R.** Aluminum and Alzheimer's disease. (1986) *Neurobiol. Aging*, 7, 525.
- Crapper McLachlan, D.R., Lukiw, W.J., & Kruck, T.P.A.** (1989) New evidence for an active role of aluminum in Alzheimer's disease. *Can. J. Neurol. Sci.*, 16, 490.
- Driscoll, C.T., Baker, J.P., Bisogni, J.J., and Schofield, C.L.** (1980) Effects of aluminum speciation on fish in dilute acidified waters, *Nature*, 284, 161.
- Driscoll, C.T.** (1984) A procedure for the fractionation of aqueous aluminum in dilute acidic waters. *Int. J. Env. Anal. Chem.*, 16, 267.
- Driscoll, C.T. and Postek, K.M.** (1995) The chemistry of aluminum in surface waters, in *The Environmental Chemistry of Aluminum*, 2<sup>nd</sup> ed., Sposito, G., Ed., Lewis Publishers, New York, Chapter 9.
- Flaten, T.P., Glattre, E., Viste, A., and Søreide, O.** (1991) Mortality from dementia among gastroduodenal ulcer patients. *J. Epidemiol. Community Health*, 45, 203.
- U.S. General Accounting Office.** Environmental Protection: DOD Management Issues Related to Chaff 1998; GAO/NSIAD-98-219.
- Good, P.F., Perl, D.P., Bierer, L.M., and Schmeidler, J.** (1992) Selective accumulation of aluminum and iron in the neurofibrillary tangles of Alzheimer's disease: a laser microprobe (LAMMA) study. *Ann. Neurol.*, 31, 286.
- Greger, J.L., Baier, M.J.** (1983) Excretion and retention of low or moderate levels of aluminum by human subjects. *Food Chem. Toxicol.*, 21, 473.

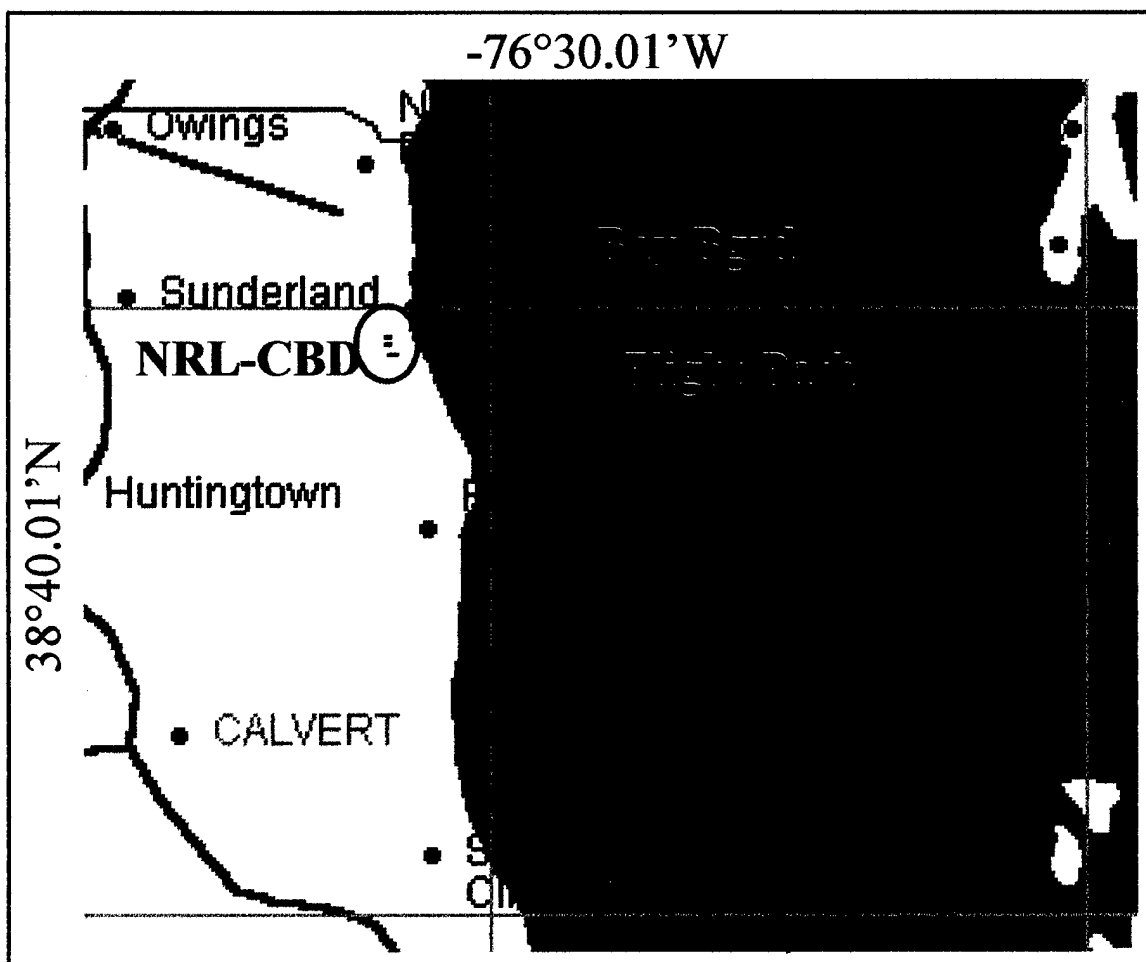
- Guy, S.P., Jones, D., Mann, D.M.A., and Itzhaki, R.F.** (1991) Human neuroblastoma cells treated with aluminum express an epitope associated with Alzheimer's disease neurofibrillary tangles. *Neurosci. Lett.*, 121, 166.
- Haley, M.V. and Kurnas, Carl W.** (1992) Aquatic toxicity and fate of iron- and aluminum-coated glass fibers, Technical Report. U.S. Army Chemical Research, Development, and Engineering Center, Research Directorate, September, Report #CRDEC-TR-422.
- Hoyt, P.B. and Turner, R.C.** (1975) Effects of organic materials added to very acid soils on pH, aluminum, exchangeable NH<sub>4</sub> and crop yields. *Soil Sci.*, 119, 227.
- Hullar, T.L., Fales, S.L., Hemond, H.F., Koutrakis, P., Schlesinger, W.H., Sobonya, R.R., Teal, J.M., and Watson, J.G.** Environmental Effects of Chaff: A Select Panel Report to the Undersecretary of Defense for Environmental Security, Naval Research Laboratory, 1999. NRL/PU/6110—99-389.
- Jouhanneau, P., Raisbeck, G.M., Yiou, F., Lacour, B., Banide, H., Drueke, T.B.** (1997) Gastrointestinal absorption, tissue retention, and urinary excretion of dietary levels of aluminum in rats as determined by <sup>26</sup>Al. *Clin. Chem.*, 43, 1023.
- Lindsay, W.L.** (1979) *Chemical Equilibria in Soils*, John Wiley & Sons, New York.
- Lindsay, W.L. and Walthall, P.M.** (1995) The solubility of aluminum in soils, in *The Environmental Chemistry of Aluminum*, 2<sup>nd</sup> ed., Spósito, G., Ed., Lewis Publishers, New York, Chapter 8.
- Martyn, C.N., Osmond, C., Edwardson, J.A., Barker, D.J.P., Harris, E.C., and Lacey, R.F.** (1989) Geographical relation between Alzheimer's disease and aluminum in drinking water. *Lancet*, 1, 59.
- Mesco, E.R., Kachen, C., and Timiras, P.S.** (1991) Effects of aluminum on tau proteins in human neuroblastoma cells. *Mol. Chem. Neuropathol*, 14, 199.
- McAvoy, D.C., Santore, R.C., Shosa, J.D., Driscoll, C.T.** (1992) Comparison between pyrocatechol violet and 8-hydroxyquinoline procedures for determining aluminum fractions. *Soil. Sci. Soc. America J.*, 56, 449.
- Neri, L.C. and Hewitt, D.** (1991) Aluminium, Alzheimer's disease, and drinking water. *Lancet*, 338, 390.
- Perl, D.P., Gajdusek, D.C., Garruto, R.M. et al.** (1982) Intraneuronal aluminum accumulation in amyotrophic lateral sclerosis and parkinsonism-dementia of Guam. *Science*, 217, 1053.

- Roberson, C.E. and Hem, J.D.** (1995) Solubility of aluminum in the presence of hydroxide, fluoride, and sulfate, U.S. Geol. Surv. Water-Supply Pap., 1827-C, 1967 in: Driscoll, C.T. and Postek, K.M. The chemistry of aluminum in surface waters, in *The Environmental Chemistry of Aluminum*, 2<sup>nd</sup> ed., Sposito, G., Ed., Lewis Publishers, New York, Chapter 9.
- Schenk, R.U., Bjorksten, J., and Yeager, L.** (1989) Composition and consequences of aluminum in water, beverages, and other ingestibles. In: Lewis, T.E., ed. Environmental chemistry and toxicology of aluminum, Chelsea, MI: Lewis Publishers, Inc, 247.
- Shin, R-W, Lee, V.M.-Y., and Trojanowski, J.Q.** (1994) Aluminum modifies the properties of Alzheimer's disease PHF(tau) proteins *in vivo* and *in vitro*. *J. Neurosci.*, 14, 7221.
- Shore, D., Wyatt, R.J.** Aluminum and alzheimer's disease. *J. Nerv. Ment. Dis.*, 1983, 171, 553.
- Stumm, W. and Morgan, J.J.** (1970) *Aquatic Chemistry*, Wiley-Interscience, New York.
- Thomas, G.W. and Hargrove, W.L.** (1984) The chemistry of soil acidity, in *Soil Acidity and Liming*, 2<sup>nd</sup> ed., Adams, F., Ed., Agronomy Series No. 12, American Society of Agronomy, Madison, WI, Chapter 1.
- U.S. Air Force Air Combat Command** (1997) Environmental Effects of Self-Protection Chaff & Flares, U.S. Air Force Air Combat Command, August.
- World Health Organization.** (1997) World Health Organization Environmental Health Criteria. EHC 194. World Health Organization, Geneva.

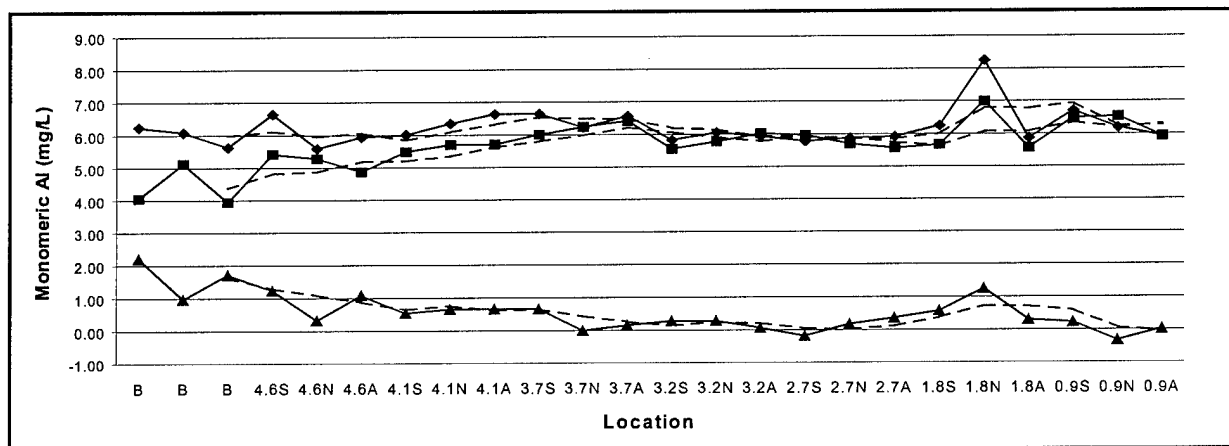


**TABLE 1. Basic constituents of chaff. Derived from Military Specification R-6034b (30).**

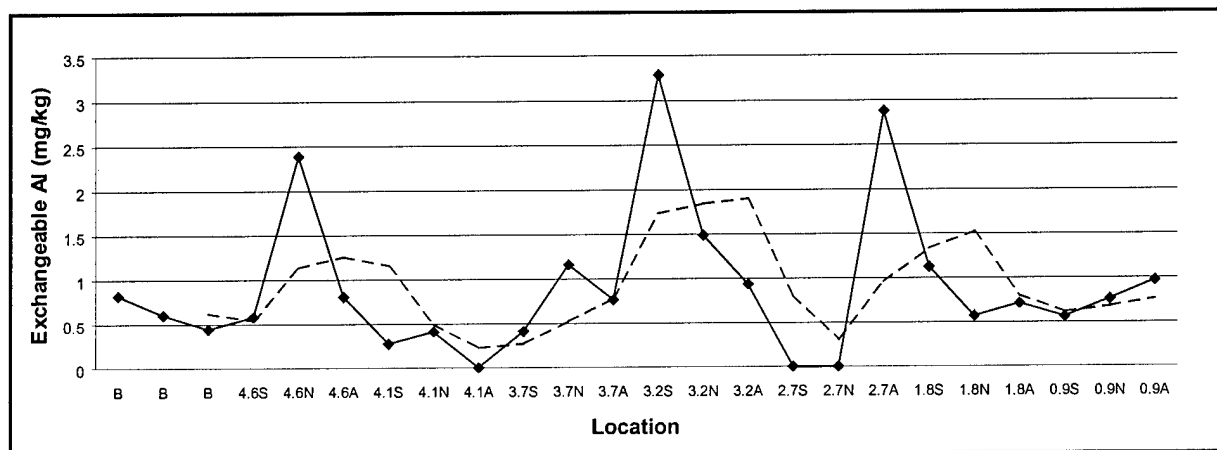
<b>Element</b>	<b>Percent by Weight</b>
<b>E-Glass Fiber</b>	
SiO <sub>2</sub>	52-56
Al <sub>2</sub> O <sub>3</sub>	12-16
CaO & MgO	16-25
B <sub>2</sub> O <sub>3</sub>	8-13
Na <sub>2</sub> O & K <sub>2</sub> O	1-4
Fe <sub>2</sub> O <sub>3</sub>	<1
<b>Aluminum coating</b>	
Al	99.45 min
Si+Fe	0.55 mix
Cu	0.05 max
Mn	0.05 max
Mg	0.05 max
Zn	0.05 max
V	0.05 max
Ti	0.03 max
Others	0.03 max



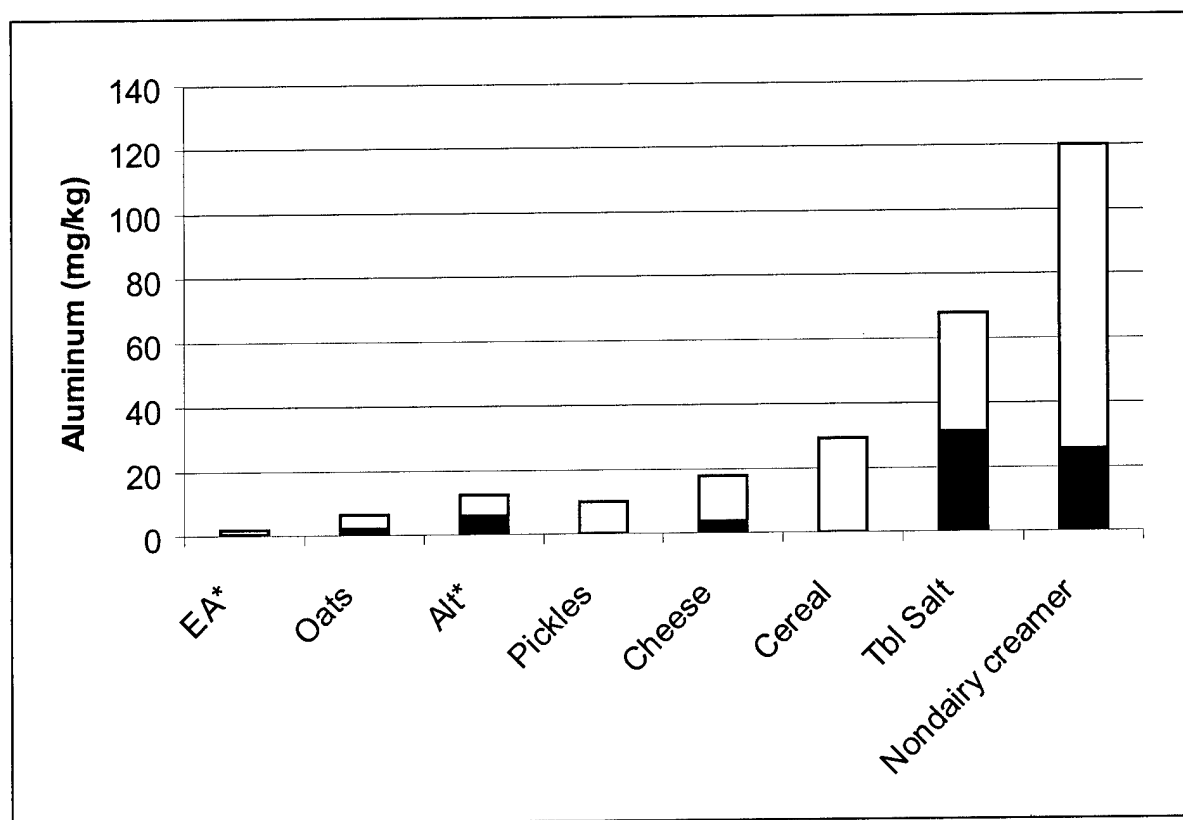
**Figure 1.** Sampling locations at NRL-CBD. Flight path and Bay background sites and sampling sites at a residential area adjacent to NRL-CBD are marked. Residential background soil was taken at an area several miles southwest of NRL-CBD (location not shown).



**Figure 2. Monomeric aluminum concentration of sediment samples obtained from a background site (B) or the Northern (N), Southern (S), or mid- (A) sites at 4600 (4.6), 4100 (4.1), 3700 (3.7), 3200 (3.2), 2700 (2.7), 1800 (1.8), and 900 (0.9) m from shore. ♦, Total monomeric aluminum; ■, organic monomeric aluminum; ▲, inorganic monomeric aluminum. The dashed lines represent a moving three point average.**



**Figure 3.** Exchangeable aluminum concentration of sediment samples obtained from a background site (B) or the Northern (N), Southern (S), or mid- (A) sites at 4600 (4.6), 4100 (4.1), 3700 (3.7), 3200 (3.2), 2700 (2.7), 1800 (1.8), and 900 (0.9) m from shore. The dashed line represents a moving three point average.



**Figure 4. Comparison of exchangeable (EA) and total monomeric (Alt) aluminum with total aluminum content of commonly consumed foods. \*, EA and Alt presented as 99% confidence intervals of all EA and Alt data collected in this study, while data from the food groups is given as a range. Alt is in mg/kg pore water. Black and white bars represent low and high values, respectively. Derived from (31).**

**TABLE 2.** Mean values for monomeric and exchangeable aluminum and pH, % solids, % total organic carbon, and exchangeable aluminum normalized to pH for samples obtained from NRL-CBD. Flight Path and Bay Bgrd samples are sediment, while CBD Res Soil and Bgrd Res Soil are soil samples. n, number of replicates; \*, sig. diff. from Bgrd Res Soil ( $p<0.05$ ); †, sig. diff. from CBD Res Soil ( $p<0.05$ ).

	<u>Flight Path</u>	<u>Bay Bgrd</u>	<u>CBD Res Soil</u>	<u>Bgrd Res Soil</u>
<b>Monomeric (mg/L)</b>				
<b>Total (Alt)</b>	6.25±0.56	5.99±0.32	na	na
<b>Org (Alo)</b>	5.86±0.48	4.37±0.66	na	na
<b>Inorg (Ali)</b>	0.39±0.43	1.62±0.62	na	na
<b>Exchangeable (mg/kg)</b>	1.10±0.87*	0.62±0.19*	27.62±43.56	47.80±19.55
<b>pH</b>	7.23±0.24*†	7.69±0.15*†	5.89±0.50	5.43±0.22
<b>% solids</b>	25.29±9.50 *†	25.33±2.49 *†	94.6±5.5	92.3±2.5
<b>TOC(%)</b>	3.01±0.64*	3.10±0.10*	1.31±0.88	0.54±0.12
<b>Exchangeable/pH</b>	0.42±0.09*	0.40±0.02*	5.15±8.22	7.14±0.50
<b>n</b>	21	3	3	5

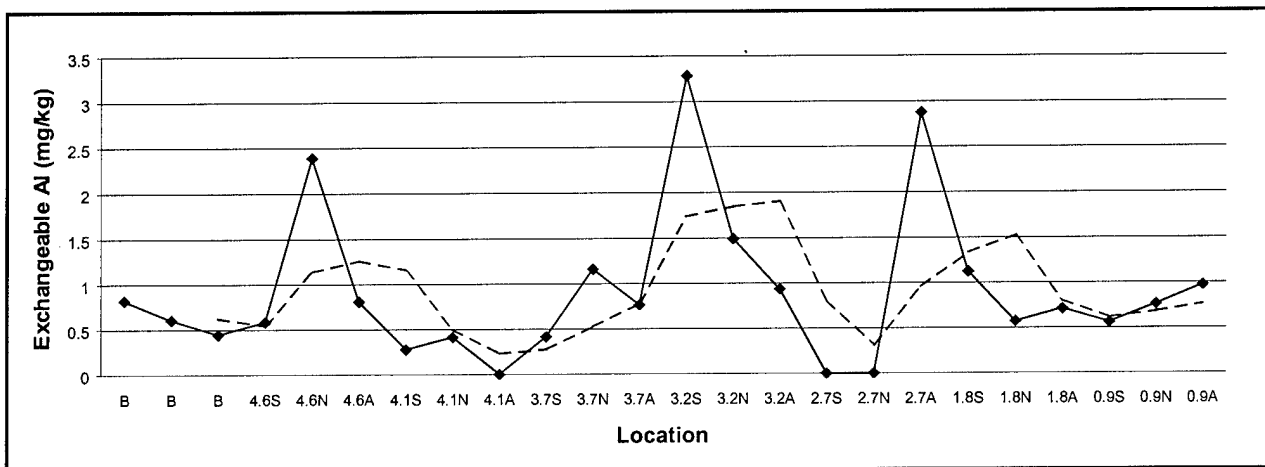


Figure 5.

<b>REPORT DOCUMENTATION PAGE</b>			<i>Form Approved</i> <b>OMB No. 0704-0188</b>	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188) Washington, DC 20503.				
<b>1. AGENCY USE ONLY (Leave blank)</b>		<b>2. REPORT DATE</b> June 2000	<b>3. REPORT TYPE AND DATES COVERED</b> June 2000	
<b>4. TITLE AND SUBTITLE</b> Estimation of Aluminum Contributions of U.S. Navy Flight Training Operations in the Chesapeake Bay			<b>5. FUNDING NUMBERS</b>  63706N-M00095.004.1822	
<b>6. AUTHOR(S)</b> C. L. Wilson, A. Miladi, R.L. Carpenter, W.K. Alexander, and K.R. Still				
<b>7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)</b> Naval Health Research Center Detachment Toxicology NHRC/TD 2612 Fifth Street, Building 433 Area B Wright-Patterson AFB, OH 45433-7903			<b>8. PERFORMING ORGANIZATION REPORT NUMBER</b>	
<b>9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)</b> Naval Health Research Center Detachment Toxicology NHRC/TD 2612 Fifth Street, Building 433 Area B Wright-Patterson AFB, OH 45433-7903			<b>10. SPONSORING/MONITORING AGENCY REPORT NUMBER</b>  TOXDET-00-04	
<b>11. SUPPLEMENTARY NOTES</b>				
<b>12a. DISTRIBUTION AVAILABILITY STATEMENT</b>  Approved for public release; distribution is unlimited.			<b>12b. DISTRIBUTION CODE</b>	
<b>13. ABSTRACT (Maximum 200 words)</b> The U. S. Navy currently uses aluminized glass chaff as a passive countermeasure for radar-guided threats to both aircraft and surface ships. Chaff countermeasures are composed primarily of aluminized glass strands with lengths of up to 2 inches and a diameter of approximately 1 mil. Until recently, research and development efforts and production lot testing of chaff countermeasures have been conducted primarily at the Naval Research Laboratory—Chesapeake Bay Detachment (NRL-CBD) near Chesapeake Beach, MD. Chaff has been deployed in training and production lot testing in this region of the Bay for nearly 25 years, suggesting a potentially significant contribution to soil and sediment aluminum levels. This study examined the exchangeable and monomeric aluminum content of bay sediment and soil from a nearby residential area to address concerns over the environmental and potential human health impact of deployed chaff countermeasures. The results suggest that sediment monomeric and exchangeable aluminum levels in the NRL-CBD testing flight path area are not significantly different ( $p < 0.01$ ) from nearby background levels within the Bay. Moreover, background residential EA levels were not significantly different ( $p < 0.01$ ) from soil samples obtained from a residential area adjacent to NRL-CBD.				
<b>14. SUBJECT TERMS</b> Chaff, exchangeable aluminum, monomeric aluminum, aluminum, sediment, Chesapeake Bay.			<b>15. NUMBER OF PAGES</b> 36	
			<b>16. PRICE CODE</b>	
<b>17. SECURITY CLASSIFICATION OF REPORT</b> UNCLASSIFIED	<b>18. SECURITY CLASSIFICATION OF THIS PAGE</b> UNCLASSIFIED	<b>19. SECURITY CLASSIFICATION OF ABSTRACT</b> UNCLASSIFIED	<b>20. LIMITATION OF ABSTRACT</b> UL	



### GENERAL INSTRUCTIONS FOR COMPLETING SIF 298

The Report Documentation Page (RDP) is used in announcing and cataloging reports. It is important that this information be consistent with the rest of the report, particularly the cover and title page. Instructions for filling in each block of the form follow. It is important to *stay within the lines* to meet *optical scanning requirements*.

**Block 1. Agency Use Only** (*Leave blank*).

**Block 2. Report Date.** Full publication date including day, month, and year, if available (e.g. 1 Jan 88). Must cite at least the year.

**Block 3. Type of Report and Dates Covered.** State whether report is interim, final, etc. If applicable, enter inclusive report dates (e.g. 10 Jun 87 - 30 Jun 88).

**Block 4. Title and Subtitle.** A title is taken from the part of the report that provides the most meaningful and complete information. When a report is prepared in more than one volume, repeat the primary title, add volume number, and include subtitle for the specific volume. On classified documents enter the title classification in parentheses.

**Block 5. Funding Numbers.** To include contract and grant numbers; may include program element number(s), project number(s), task number(s), and work unit number(s). Use the following labels:

C - Contract	PR - Project
G - Grant	TA - Task
PE - Program Element	WU - Work Unit Accession No.

**Block 6. Author(s).** Name(s) of person(s) responsible for writing the report, performing the research, or credited with the content of the report. If editor or compiler, this should follow the name(s).

**Block 7. Performing Organization Name(s) and Address(es).** Self-explanatory.

**Block 8. Performing Organization Report Number.** Enter the unique alphanumeric report number(s) assigned by the organization performing the report.

**Block 9. Sponsoring/Monitoring Agency Name(s) and Address(es).** Self-explanatory.

**Block 10. Sponsoring/Monitoring Agency Report Number.** (If known)

**Block 11. Supplementary Notes.** Enter information not included elsewhere such as: Prepared in cooperation with ....; Trans. of ....; To be published in.... When a report is revised, include a statement whether the new report supersedes or supplements the older report.

**Block 12a. Distribution/Availability Statement.** Denotes public availability or limitations. Cite any availability to the public. Enter additional limitations or special markings in all capitals (e.g. NOFORN, REL, JTAR).

**DOD** - See DoDD 5230.24, "Distribution Statements on Technical Documents.

**DOE** - See authorities.

**NASA** - See Handbook NH13 2200.2.

**NTIS** - Leave blank.

**Block 12b. Distribution Code.**

**DOD** - Leave blank.

**DOE** - Enter DOE distribution categories from the Standard Distribution for Unclassified Scientific and Technical Reports.

**NASA** - Leave blank.

**NTIS** - Leave blank.

**Block 13. Abstract.** Include a brief (*Maximum 200 words*) factual summary of the most significant information contained in the report.

**Block 14. Subject Terms.** Keywords or phrases identifying major subjects in the report.

**Block 15. Number of Pages.** Enter the total number of pages.

**Block 16. Price Code.** Enter appropriate price code (*NTIS only*).

**Blocks 17. - 19. Security Classifications.** Self-explanatory. Enter U.S. Security Classification in accordance with U.S. Security Regulations (i.e., UNCLASSIFIED). If form contains classified information, stamp classification on the top and bottom of the page.

**Block 20. Limitation of Abstract.** This block must be completed to assign a limitation to the abstract. Enter either UL (unlimited) or SAR (same as report). An entry in this block is necessary if the abstract is to be limited. If blank, the abstract is assumed to be unlimited.